

Commercial Light Water Production of Tritium: Update and Path Forward

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Tritium Focus Group



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Background

- ▶ United States defense maintains a stockpile of nuclear weapons as a “deterrent” to military actions by others
- ▶ Tritium is required for all U.S. nuclear weapons to function as designed: ${}^6\text{Li} + {}^1_0\text{n} \rightarrow {}^3_1\text{T} + {}^4_2\text{He}$
- ▶ With a 12.2 year half-life, tritium must be replaced. Department of Energy (DOE) stopped production of tritium at Savannah River Site (SRS) in 1988.
- ▶ 1988 – 1992: The U.S. considered use of dedicated reactors; heavy water reactors (SRS), high temperature reactors (Idaho), and light water reactors (Hanford). LWR choice was WNP-1 (partially completed)
- ▶ 1992 brought the break-up of the Soviet Union and Start treaties to reduce weapon stockpiles. Net result: much less tritium needed to be produced.
- ▶ U.S. considered the use of an accelerator or commercial light water reactors to meet the reduced quantities.
 - LWR was selected for production deployment.



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Tritium Program Timeline

- 1995 – DOE Record of Decision for dual track approach to produce tritium
- 1997 – Initial irradiation of Lead Test Assembly (LTA)
(32 Tritium Producing Burnable Absorber rods (TPBARs))
- 1999 – Post Irradiation Examinations (PIE) of LTA rods
- 2003 – First Production Operations (240 TPBARs)
- 2004 – Determine (from coolant analysis) that tritium production permeation into reactor coolant higher than predicted.
 - Models predicted .5 Ci/Rod/Year
 - Actual Permeation would not allow desired amount of tritium to be produced at current allowable release limits.
- 2004-2007 –Redesign to mitigate permeation and address manufacturing lessons learned

Tritium Program Timeline (cont.)

2008 – Irradiation of new design

2009 – Confirmation that re-design did not improve permeation: emphasis shifts to Demonstration and Testing (D&T) program using Idaho's Advanced Test Reactor (ATR)

2010 – Initial test data from D&T program; Production limited to 704 TPBARs without revision to Large Break Loss of Coolant Accident methodology

2012 – Production increased to 544 TPBARs with Lead Use Rods, Draft Environmental Impact Statement submitted for approval to increase allowable releases of tritium, “Big Tank” designed and installed at TVA

WBN1 TPBAR Irradiation Schedule

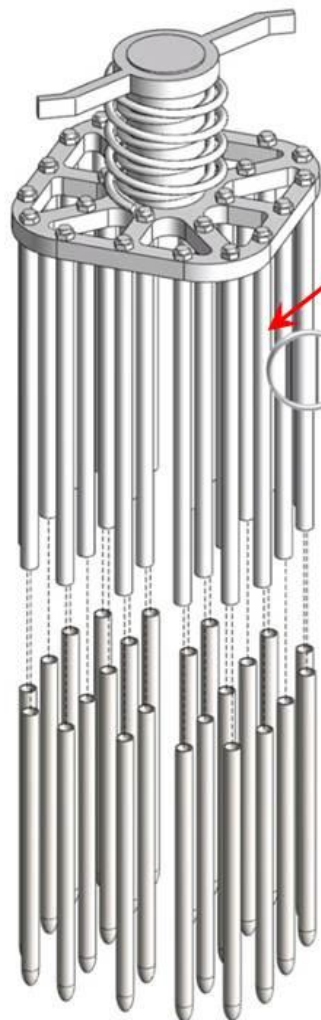
WBN Cycle	# of TPBARs in Cycle	Cycle Start Date	Cycle End Date
Cycle 6	240	2003-Sept	2005-Feb
Cycle 7	240	2005-Mar	2006-Sept
Cycle 8	240	2006-Nov	2008-Feb
Cycle 9	368	2008-Mar	2009-Sept
Cycle 10	240	2009-Oct	2011-Apr
Cycle 11	544	2011-May	2012-Oct
Cycle 12	544	2012-Nov	2014-Apr
Cycle 13	704	2014-May	2015-Oct
Cycle 14	1104*	2015-Nov	2017-Apr
Cycle 15	1504*	2017-May	2018-Oct
Cycle 16	1696*	2018-Nov	2020-Apr

*actual numbers depend on core analysis and tritium production goals

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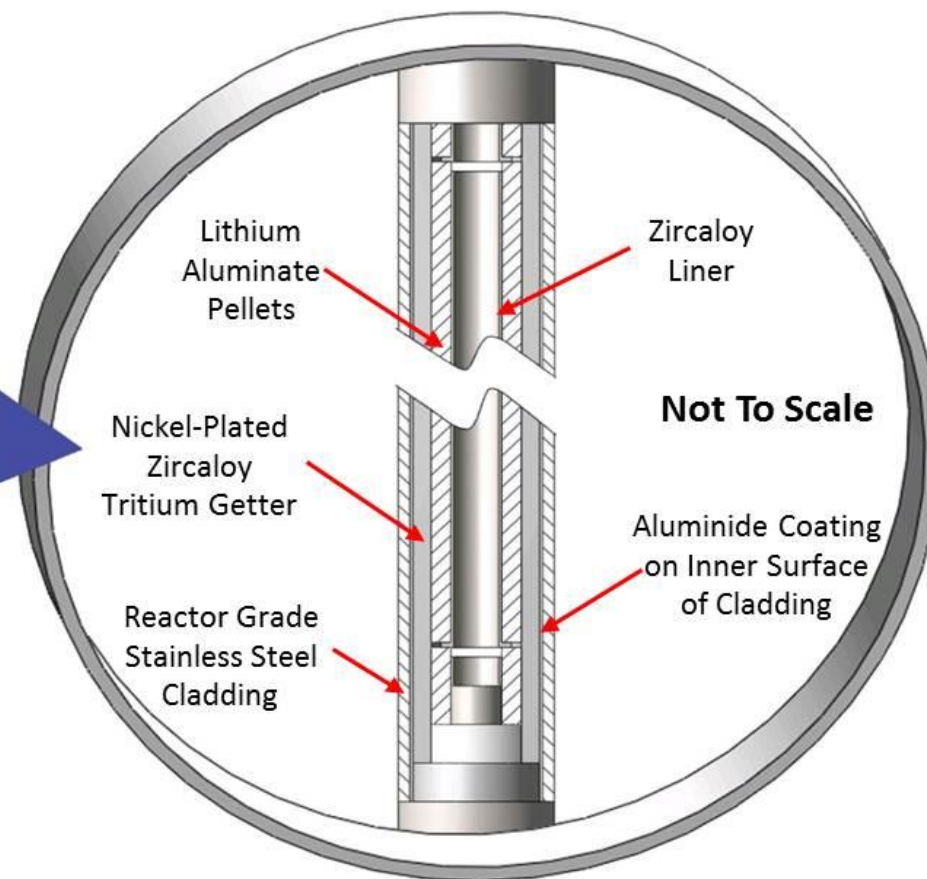
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Tritium Producing Burnable Absorber Rod (TPBAR)



Assemblies consist of 12 to 24 rods suspended from a base plate

Each rod is about 12.5 feet long and $\sim \frac{3}{8}$ inch in diameter



Functions of TPBAR Components

Stainless Steel Cladding – Similar to reactor fuel elements. Contains all TPBAR components

Aluminide Coating – Limits permeation of tritium through the stainless steel cladding into the reactor coolant. Also limits hydrogen in the coolant from entering the TPBAR.

Zircaloy (zirconium alloy) Tritium Getter – Absorbs free tritium gas

Nickel Plating – Prevents oxidation of the tritium getter

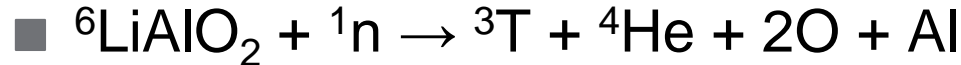
Lithium Aluminate Pellets – High temperature ceramic material containing lithium-6, the material that transmutes to tritium when a neutron is absorbed

Zircaloy Liner – Reduces tritiated water so the tritium can be absorbed by the getter.

During and after irradiation, nearly all of the tritium is retained in the ceramic, the tritium getter and the Zircaloy liner until it is released during the extraction process. There is little free tritium in the gas.

General Tritium Processes within a TPBAR

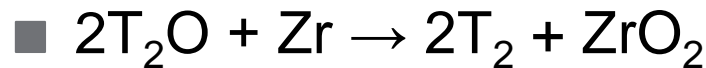
▶ Neutron capture



▶ Release from the pellet



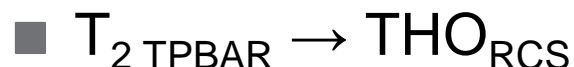
▶ Cracking on the liner and cruciforms



▶ Absorption by the getter to reduce the partial pressure of T_2



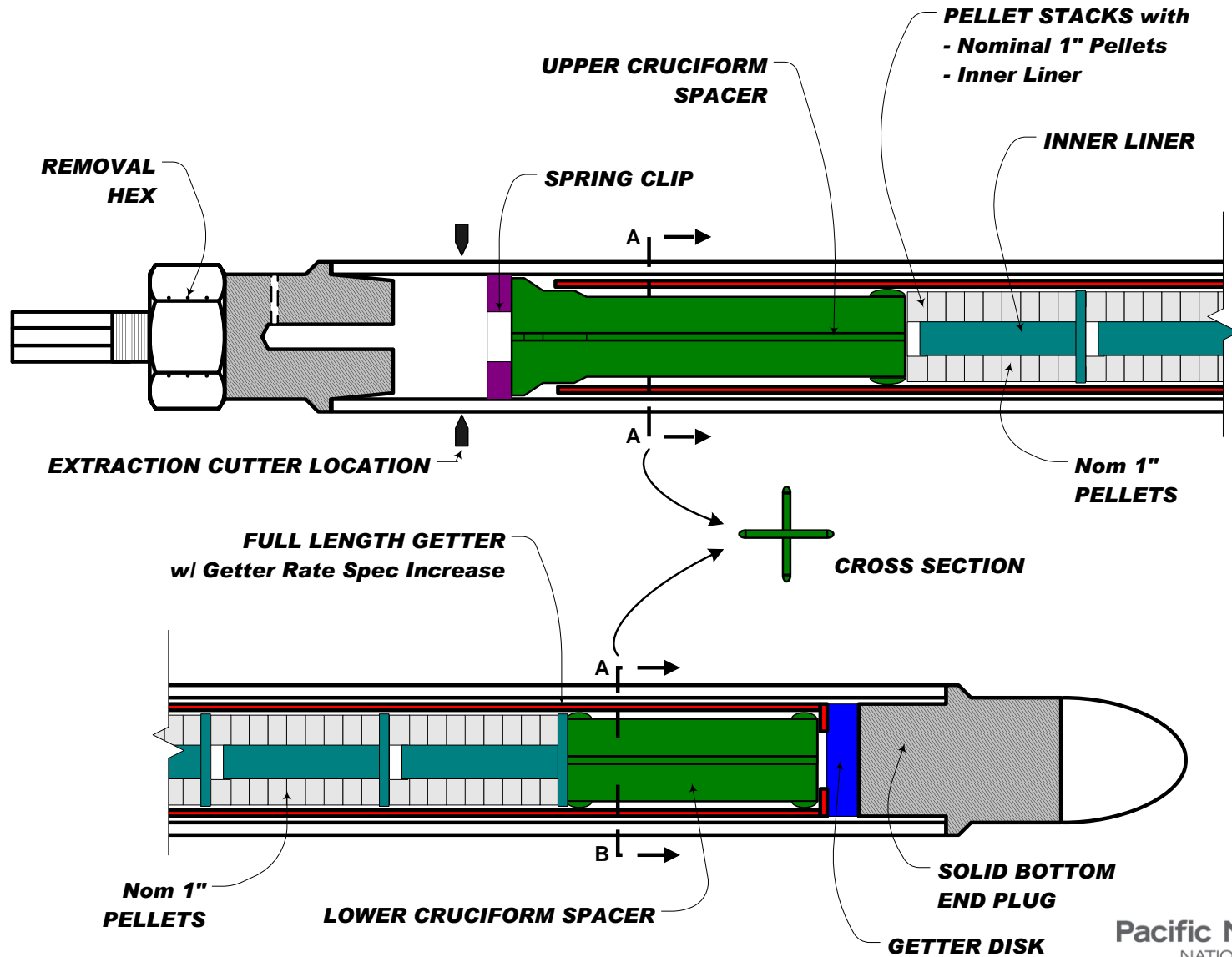
▶ Tritium permeation into the reactor coolant system (RCS)



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Mark 9.2 - Configuration



TPBAR Irradiation Performance

- ▶ In 2004, during Cycle 6, it was determined that TPBAR tritium permeation was higher than predicted by performance models
 - Predicted ≈ 0.5 Ci/TPBAR/cycle
 - Actual ≈ 4 Ci/TPBAR/cycle
- ▶ Even 4 Ci/TPBAR/cycle represents only about 0.04% of the tritium produced
- ▶ TVA limited the number of TPBARs that could be irradiated because of current license limits on tritium release
- ▶ Subsequent irradiations have continued, but quantities are limited to <704 TPBARs/cycle (LBLOCA issue, though permeation is a limit)
- ▶ A research and development program was implemented in 2006 to provide a scientific basis for improving performance models and providing systematic, long-term TPBAR design evolution



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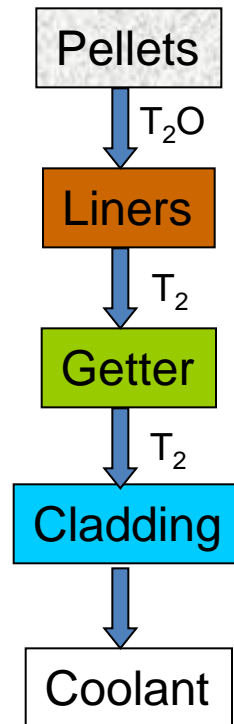
TPBAR Design Evolution

- ▶ Cycle 6 demonstrated original vision was inadequate
- ▶ PIE and ex-reactor testing led to revised vision that drove Mark 9.2 design
- ▶ Subsequent TPBAR irradiation and test reactor experiments showed Mark 9.2 vision is still inadequate

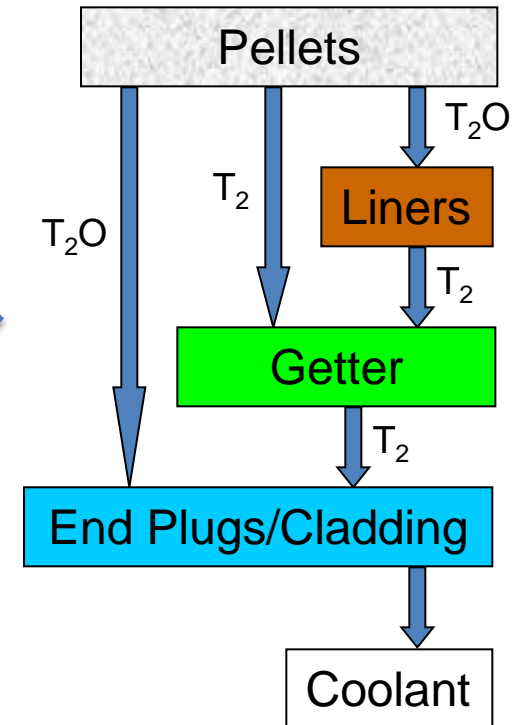
- Pellet speciation and burnup dependencies
- Carbon transport and deposition
- Tritium transport and distribution

Tritium Transport within TPBAR

Original Vision



Mark 9.2 Vision



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Principle Findings from Tritium R&D Program

- ▶ **TMIST-1/TMED-1 – Linear Oxidation & Hydrogen Uptake**
 - Quantified irradiation enhancement in Zircaloy -2 & -4 oxidation at low D₂O vapor pressure (~2X)
 - Quantified hydrogen (deuterium) uptake in Zircaloy -2 & -4 in reactor (10-40%), irradiation enhancement of 1-4X
 - Evaluated surface-modified Zircaloy-4 for oxidation kinetics & deuterium uptake
 - Observed differences between separate-effects and integral environments
- ▶ **TMIST-2 – Stainless Steel Permeation of Tritium**
 - Quantified irradiation enhancement in 316SS for tritium permeation (~2-3X)
 - Confirmed pressure dependence of tritium permeation through 316SS of $P^{0.5}$, indicating diffusion-limited permeation rate
 - No fluence effect up to $\sim 5 \times 10^{21}$ n/cm²



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Principle Findings (Cont.)

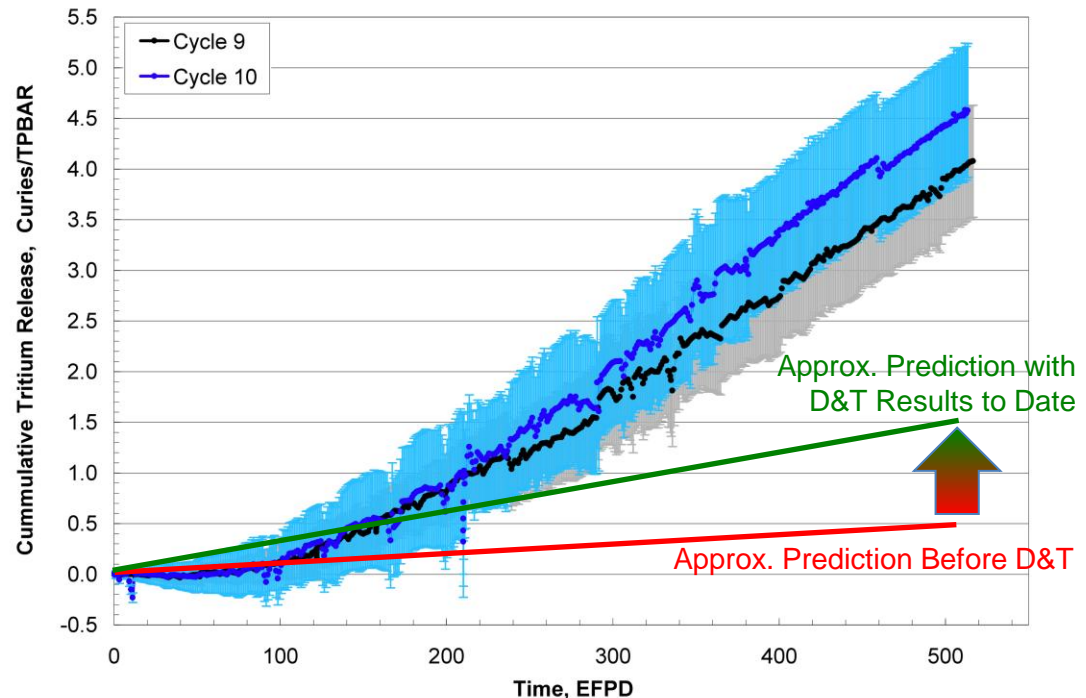
- ▶ TMED-4 – Hydrogen Partial Pressure over Ni-plated Zircaloy Getters
 - Observed differences between literature data for H pressure over Zr and H pressure over Ni-plated Zircaloy-4
- ▶ TMED-3 – Advanced Pellet Fabrication Demonstration
 - Developed LiAlO_2 pellet manufacturing processes to produce large-grain and porous pellets
 - Demonstrated engineered porosity with different sizes and morphologies
 - Developed LiAlO_2/Zr cermet pellets in LiAlO_2 volume fractions from 10 to 40%



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Data from the Testing Program Has Improved TPBAR Performance Predictions

- ▶ TROD performance prediction code models updated with data from TMIST-1, TMED-1, TMIST-2, and TMED-4
- ▶ Discrepancy between predicted and observed permeation decreased by ~30%
- ▶ Time dependence still not correctly modeled
 - Would be improved by TMIST-3 data

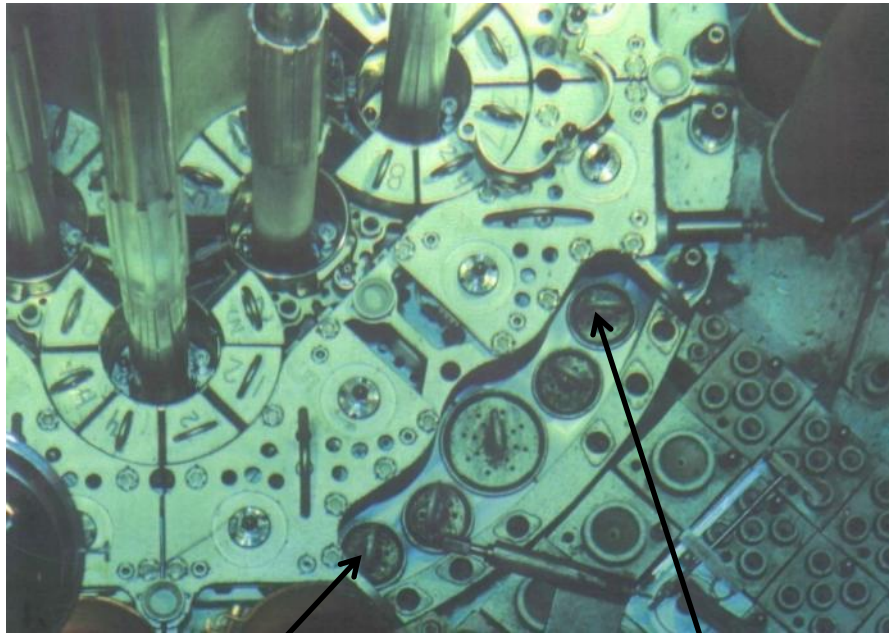


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Pellet Performance Irradiation Experiment TMIST-3

- ▶ Data from TMIST-3 will
 - Explain time dependence of pellet tritium release and its relationship to TPBAR permeation
 - Evaluate the speciation of tritium release as a function of burnup, burnup rate, and time (T_2O versus T_2)
 - Define relationships between pellet burnup, burnup rate, and tritium release to help define an acceptable TPBAR operational envelope
 - Improve fundamental understanding of pellet microstructure and its effects on performance
 - Provide a better definition of the pellet burnup limit
 - Determine whether modifications to the pellets could improve TPBAR performance
 - Increased tritium retention
 - Increased TPBAR void volume

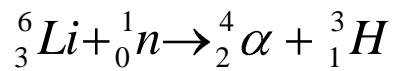
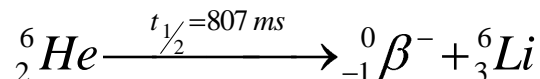
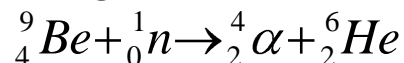


Location for the TMIST-3A
low-burnup test train (I-13)

Location for the TMIST-3B
high-burnup test train (I-9)

TPBAR Performance History

- ▶ In 2007, PNNL began work to better understand non-TPBAR tritium sources in the coolant
 - Reactions from boron, lithium, and ternary fission sources in fuel were all known
 - Fuel releases were determined to be minimal with Zircaloy clad
- ▶ Secondary sources turned out to be a major contributor of tritium to the coolant (for determining TPBAR permeation)
- ▶ Tritium is generated from the secondary sources according to the reactions



- Cycles had 1, 2 or 3 sources with various irradiation histories

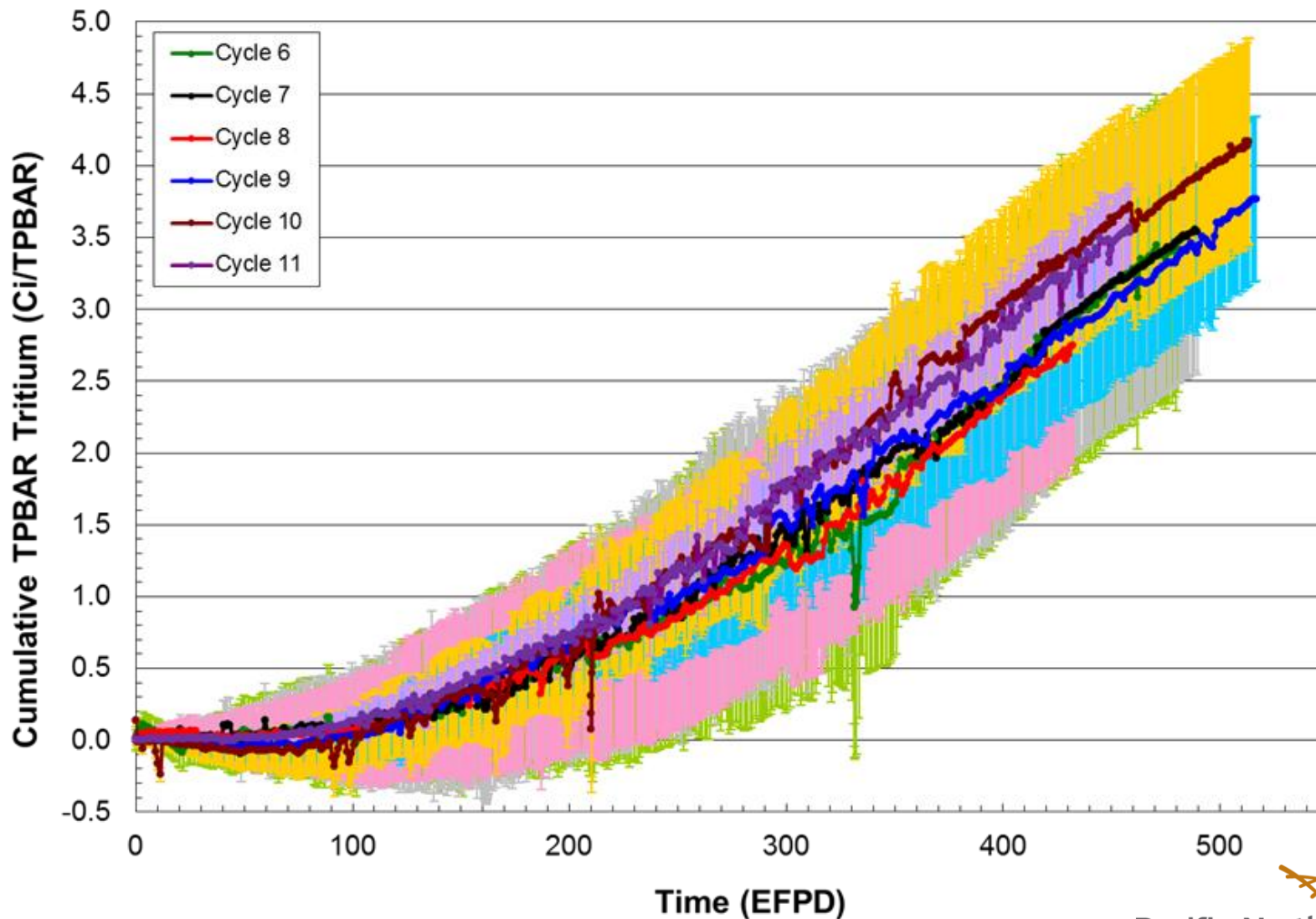


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TPBAR Performance History (cont.)

- ▶ In 2009, PNNL began revising all TPBAR tritium estimates accounting for secondary source contributions and removing constant fuel release assumption
- ▶ Accounting for secondary sources:
 - Cycle 6-8 TPBAR releases have been re-evaluated to be 3.7 ± 1.0 Ci/rod/year, 3.2 ± 0.9 Ci/rod/year, 3.1 ± 0.8 Ci/rod/year, respectively (3.3 ± 0.6 Ci/rod/year average)
 - Cycle 9 TPBAR release is estimated at 3.5 ± 0.6 Ci/rod/year
 - Cycle 10 TPBAR release is estimated at 3.8 ± 0.8 Ci/rod/year
 - Cycle 11 TPBAR release is estimated at 3.4 ± 0.5 Ci/rod/year

Estimated TPBAR Tritium Release For WBN1 Cycles 6 Through 10



Performance Modeling Progress

- ▶ The TPBAR permeation predictions have become significantly closer to TPBAR release estimates derived from coolant measurements
 - Previous prediction was ~ 0.5 Ci/rod/yr
 - Current prediction ~ 1.5 Ci/rod/yr
- ▶ Conclusion:
 - It is extremely important to have a firm understanding of the predictive equation parameters used
 - 2nd, 3rd, and possibly 4th order effects may have a significant impact in this range but are not well understood
 - Designers MAY have a complete understanding of mechanisms that effect TPBAR performance, but an inadequate understanding of the equation's coefficients
 - Or there may still be a mechanism that we are missing



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Conclusions

- ▶ Current design is very efficient
- ▶ Actual improvements in permeation may be very difficult to achieve
- ▶ However, understanding permeation is very important to the program
 - 50 year program life
 - Reactor operating changes will occur
 - Manufacturing changes will occur
 - Management of tritium releases very important to the program: no surprises!



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